Optical Characterization of Lateral Epitaxial Overgrown GaN Layers

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The optical properties of homoepitaxial GaN layers deposited by organometallic vapor phase epitaxy on stripe-patterned GaN films on 6H-SiC substrates have been investigated. Analysis of the spatially-resolved Raman scattering spectra indicate an improvement in material quality of the overgrown region. Room-temperature color cathodoluminescence imaging and low-temperature photoluminescence measurements indicate that a donor and an acceptor, different from those detected in the underlying GaN/AlN/SiC substrate, have been incorporated in the epitaxial layer. Detailed photoluminescence studies of the near band-edge emission strongly suggest that Si is the additional donor detected in the homoepitaxial GaN layer. Its occurrence, along with that of an acceptor-related defect which is primarily found in the laterally overgrown region, is discussed.

Recently it was demonstrated that the usual high threading dislocation density in heteroepitaxial layers can be significantly reduced using a patterned GaN film for selective homoepitaxial deposition [1,2]. In this work we report the optical and electronic properties of GaN layers selectively overgrown by metalorganic vapor phase epitaxy (MOVPE) on GaN/AlN/6H-SiC substrates.

Each of the initial 1.5-2.0 μm thick GaN films was deposited on a 100 nm AlN buffer layer prepared by deposition on 6H-SiC(0001) substrates [3]. These films were subsequently covered with a 100 nm SiO_2 film which were patterned by standard lithography and chemical etching [2]. The stripe patterns included both 3 μm and 5 μm wide stripe openings oriented along $\langle 1\bar{1}00 \rangle$ and etched through the SiO_2 film. Different areas had different stripe separations which range from 3 μm to 40 μm. The patterned substrates were reinstalled in the MOVPE reactor. The amount of vertical and lateral epitaxial overgrowth (LEO) showed a strong dependence on stripe orientation and growth conditions [5]. Under optimized growth conditions rectangular cross-sectional stripes were deposited on both 3 μm and 5 μm wide stripes [2]. Cross-sectional and plan view transmission electron microscope TEM micrographs of isolated stripes show that the threading dislocations originating at the GaN/AlN interface are confined to the vertical growth region (window regrowth: WR) [2]. Continuous films were achieved for both 3 μm and 5 μm wide stripes; however, we will present and discuss only the results related to isolated stripes where adjacent overgrowth regions had not coalesced.

The samples were initially characterized by spatially-resolved room temperature Raman scattering (RS) using the 514 nm line of an Ar ion laser focused to a 1 μm spot size [4]. The 325 nm line of a HeCd laser which was incident either normal to or at 60 degrees from normal was the exciting source for the low-temperature PL experiments. An 0.85 m double-spectrometer with 1800 gr/mm gratings, a low noise photomultiplier, and a photon counting system were used to analyze the light emitted by the samples [5]. The PL experiments were conducted with comparable or higher spectral resolution than the RS

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measurements; however, the spot size of the PL measurements was either ~20μm or ≥60 μm. Color-cathodoluminescence scanning electron microscopy (CCL-SEM, room temperature) [6] was used to investigate the spatial distribution of defects associated with emission bands observed in the PL spectra.

The RS spectra of individual stripes for both sample orientations, planar and cross-sectional view, show all phonons allowed by selection rules [5,7]. A small peak shift and a ~20% reduction of the linewidth of the E_2 phonon was observed for the LEO region compared with that found in the GaN film away from the selectively grown region. These observations are consistent with the reduction of biaxial strain and structural defect concentration in the LEO region [7].

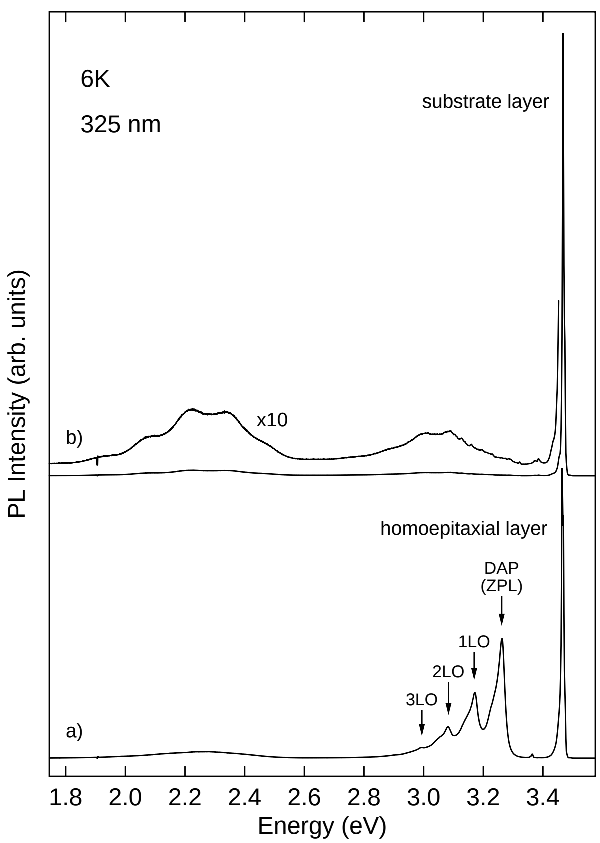

Fig 1. Low resolution PL spectrum of (a) the 3μm uncoalesced stripes and (b) the GaN/6H-SiC substrate.

Low temperature PL spectra excited with a ≥60 μm diameter laser beam incident on a region of the sample where LEO had developed from 3μm wide stripes are presented in spectrum "a" of Fig. 1. A relatively weak yellow band (2.25 eV), the edge emission band (3.467 eV), and an intense donor-acceptor pair (DAP) band with zero phonon line (ZPL) at 3.263 eV and with phonon replicas at 3.173 eV, 3.083 eV, and 2.992 eV were observed. The PL spectrum of the GaN/AlN/SiC substrate (spectrum "b" in Fig. 1) does not show evidence of the 3.263 eV DAP band. However, a deeper featureless band centered around 3.05 eV which is commonly detected in films with high-field breakdown was observed [8]. Although these results indicate that the defect associated with the DAP band is present in the homoepitaxial GaN layers, the low spatial resolution of the PL measurements prevents us from establishing whether these defects are incorporated in the WR or in the LEO region.

To investigate the distribution of radiative defects across the homoepitaxial layer we performed CCL-SEM imaging experiments. The data from uncoalescent-stripe regions in two different samples is shown in Fig. 2. Fig. 2a shows the CCL image of homoepitaxial stripes grown through 5 μm windows separated by 25 μm. In this case nonoptimized growth conditions led to the formation of homoepitaxial layers of trapezoidal cross section. Note that the fast vertical growth rate resulted in the incorporation of a high density of defects associated with yellow emission. However, blue emission is clearly observed on the triangular-shaped LEO region. Rectangular cross-sectional epitaxial stripes have been prepared by controlling the vertical growth rate. The CCL image of homoepitaxial layers grown in 3 μm wide windows separated by 12 μm is shown in Fig. 2b. Although most of the yellow emission originates from the region between the stripes, where the underlying GaN film is also excited by the e-beam, a low intensity yellow emission is observed in the WR region. The planar-CCL imaging of the region in Fig. 2(b) at one half magnification is represented in Fig. 2©. The stripes show a dominant blue color because of the higher intensity and light-piping of the blue emission from the LEO region. The green emission observed between the stripes in Fig. 2©

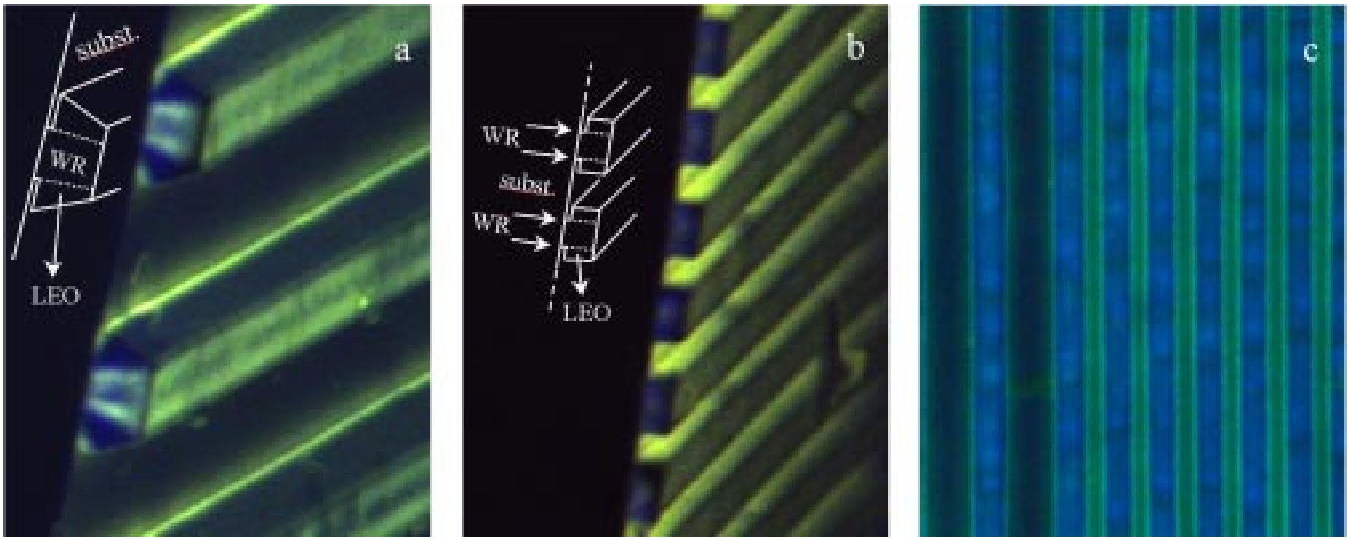

Fig. 2. Full-color CL imaging of uncoalesced stripes. (a) 5 μm wide window region. Non-optimized growth condition induced the trapezoidal cross-sectional morphology, with threading dislocations extending in to the LEO region. (b) 3 μm wide window region. Optimized growth condition resulted in a rectangular cross-sectional morphology and confinement of the threading dislocation within the window regrown (WR) regions. The recombination process associated with the blue emission band becomes dominant at the LEO region. (c) Planar CL imaging of the 3 μm wide-windows represented in (b). Note that the magnification in (c) is one half of that in (a) and (b). The horizontal field of view in (a) and (b) is 50 μm. Schematic diagrams representing the LEO and WR regions for trapezoidal and rectangular morphological growth are included in (a) and (b), respectively.

results from the mixing between the blue and the yellow emissions originating from the LEO and the underlying GaN film, respectively.

Higher spectral resolution measurements of the bandedge emissions were performed to identify the impurities associated with the PL signatures. The PL spectrum of the underlying GaN film shows two peaks which have been assigned to the free exciton A (FX_A; ~3.4725 eV) and to an exciton bound to a neutral donor ($X\text{-}D^0$; ~3.4667 eV) [9]. The PL spectra of all uncoalesced stripes show an additional band at ~3.4643 eV. In these measurements the laser probing spot was about 10 times larger than one individual stripe (~6 μm), and the angle of incidence was about 60° from normal to the sample surface. Therefore, for large stripe separations both homoepitaxial stripes as well as the underlying GaN film were probed. By moving the laser spot to regions with smaller stripe separation, the underlying GaN film contribution was reduced with respect to the stripe contribution. Under these conditions sharper and blue-shifted emission bands were observed. These observations are consistent with improved GaN material quality and/or a more relaxed, strain-free environment in the LEO region. However, the nature of the additional line at ~3.4643 eV was not explained. To determine whether this line results from a second donor or from the same donor in the highly-strained (WR) region, we carried out a variable temperature experiment in a region where both lines show about the same relative strength. The results are summarized in Fig. 3. The spectra show the expected systematic reduction of the PL band intensities from 6K to 50K (curve “f”). Comparison between the 50K spectrum of the overgrown GaN layers with the 50K spectrum (curve “g”) of the underlying GaN film indicated that the additional line has a larger residual intensity than the pervasive donor line. Therefore this additional band should be associated with excitons bound to a presumably deeper second donor.

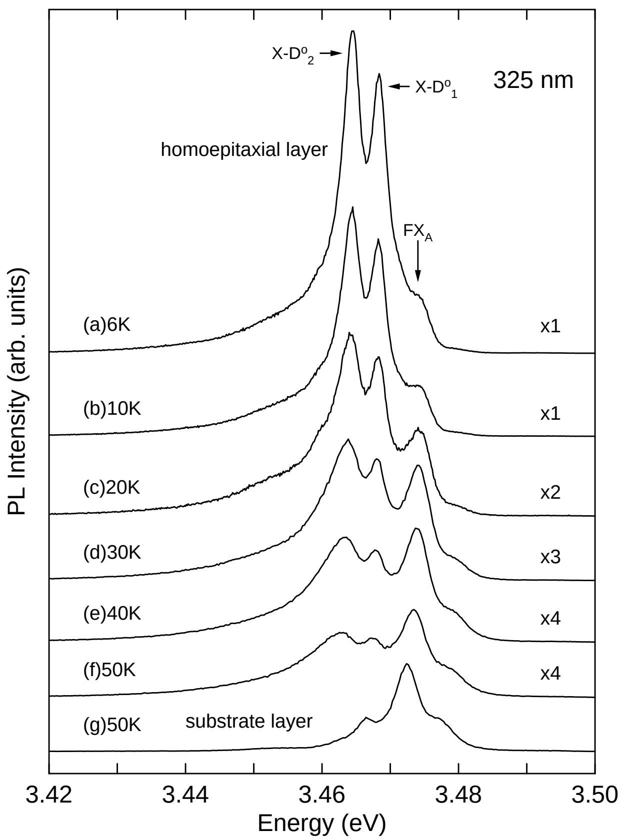

Fig. 3. Near band-edge PL spectra of 3µm uncoalesced stripes at different temperatures. Note the of the absence of the line at 3.4643 eV (D_2) in curve (g); the 50K spectrum of the GaN substrate.

Low temperature and high spectral resolution PL spectra obtained with a ~20 µm laser spot in a region of almost coalesced stripes and in a region where only the underlying GaN film was present are presented in Fig. 4. A dominant exciton bound to the second donor (X-D_2) in the PL spectrum of the homoepitaxial layers was observed, in addition to the pervasive donor (D_1) and the free-exciton related features blue-shifted by ~1.5 meV. The separation between D_2 and FX_A is about 9.6 meV, while that between D_1 and FX_A is about 5.6 meV. Since we have used SiO_2 for substrate masking, and the regrowth was carried out at high growth temperatures, it would not be surprising to find oxygen and/or silicon incorporated in the film. The spectral position is not conclusive evidence of an impurity identity; however reported PL studies of undoped and Si-doped GaN films [8] suggest that Si is the second donor detected in the homoepitaxial layers. On that basis, a smaller feature around 3.4505 eV is tentatively assigned to an oxygen-related defect. Additional experiments such as secondary ion mass spectroscopy (SIMS) will be conducted to verify these assignments. The Si and O related lines were not observed in regions where the regrown stripes coalesced. This observation indicates that the incorporation of O and Si stop upon coalescence, as the SiO_2 is now buried under the continuous GaN overgrown layer. The broadening and red-shift of the band edge emission suggests that a large biaxial strain is present in the coalesced layer.

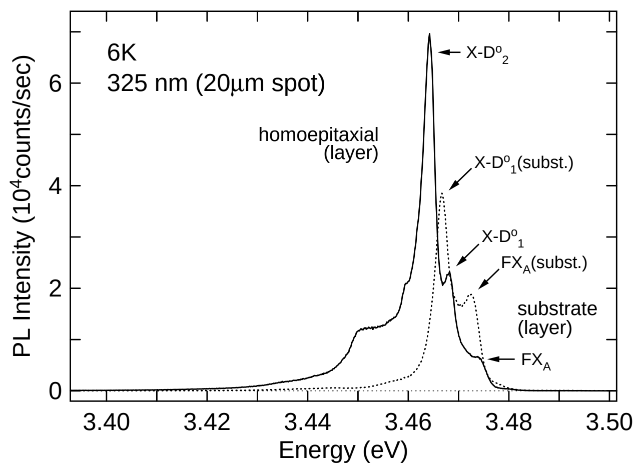

Fig. 4. Low temperature higher spectral resolution PL spectra from 3µm uncoalesced stripes and GaN substrate regions. The laser spot was ~20 µm. Note the new features at ~3.4506 eV and 3.4642 eV, and the relative blue-shift (~1.5 meV) of D_1, FX_A, and FX_B in the overgrown GaN spectrum.

The optical and electronic properties of homoepitaxial selectively overgrown stripes with $\{11\bar{2}0\}$ and $\{1\bar{1}01\}$ side facets deposited by MOVPE on GaN/AlN/6H-SiC substrates have been presented here. Spatially-resolved Raman scattering performed on isolated stripes are consistent with reduction of the biaxial strain and defect concentration. Low temperature PL spectra from the homoepitaxial layers exhibited an additional emission band with a ZPL at 3.263 eV, which we assigned to a recombination process involving shallow donor and acceptor defects. Data from CCL-SEM experiments indicated that this acceptor is preferentially located within the

LEO region. Detailed temperature dependence studies of the edge-emission band from the homoepitaxial GaN stripes suggested the assignment of a new band to the Si donor. Based on the Si assignment, we speculate that the additional feature is related to the presence of O.

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